An open systems approach to calculating time dependent spectra

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A new method to calculate the spectrum using cascaded open systems and master equations is presented. The method uses two state analyzer atoms which are coupled to the system of interest, whose spectrum of radiation is read from the excitation of these analyzer atoms. The ordinary definitions of a spectrum uses two-time averages and Fourier-transforms. The present method uses only one-time averages. The method can be used to calculate time dependent as well as stationary spectra.

I. INTRODUCTION

Much of the information obtained in laser experiments comes in the form of spectral data. In steady state, these are supposed to give the Fourier transform of the energy level structure of the system under investigation. In this manner we have been able to learn about the quantum configurations of atoms, molecules and solid state systems.

Today, however, much work is done with pulsed laser sources, where the pulse duration samples the evolution of the system over times ranging from nanoseconds to femtoseconds. Spectral data are still recorded, but their significance and physical information is no longer straightforward. Thus one needs to reconsider the definition and computation with due consideration of the physical conditions under which the data are obtained. Several attempts have been made to modify the Fourier-transform definition, the Wiener-Khintchine spectrum [1–3], but here we choose to consider the physical spectrum introduced by Eberly and Wódkiewicz [4], which attempts to emulate the spectral measurements using e.g a Fabry-Perot filter in front of the detector. They have applied it to the well known case of the fluorescence Mollow spectrum [5]. A very similar approach has allowed Kowalczyk et al. [6] to consider the fluorescent spectra deriving from a laser-excited molecular wave packet. A model calculation of such a process has been presented by Vinogradov and Janszky [7]. Another approach to the molecular situation is presented by Lee et al. [8].

In this paper we consider the problem of obtaining spectral data in an evolving system. Thus only the accumulated information is available; the future is still unknown. This rules out the use of a full Fourier-transform, and only physically manipulated collected data can be used. The spectral measurements require a filtering, which smeares the signal in time. The frequency-time resolution has to obey an uncertainty relation. In addition, the exact nature of the transfer of spectral information from the system investigated to the detector imposes its own limitations. There can be no unique spectral definition for time-dependent systems, but we can require that all definitions agree when infinite measurement times are available.

In a laboratory experiment, the radiation emitted from a driven system reaches the detector through a technical setup which, in addition to its function as a filter, will impose its own noise limits. The filtering action provides a noisy channel. This is, however, just the situation which is described by the term "Open system" [11]. We want to consider an open systems approach to time-dependent spectra.

A natural frequency-selective detector is a two-level atom. This will respond to radiation only within a bandwidth given by its natural linewidth. If this is small enough, the spectrum if incoming radiation is resolved with this accuracy; this mirrors the quantum fluctuations of the detector-atom decay. We also let the radiation reach the detector atoms through a noisy channel, a reservoir which can be eliminated. Thus we present a new approach to time dependent spectra, which does not seem to be directly related to the ones used earlier. The aim of the present paper is to introduce this model and compare it with the physical spectrum of Eberly and Wódkiewicz and the Wiener-Khintchine spectrum for steady state.

In Sec. II.A we introduce the system we want to investigate the spectrum of. We choose a simple one consisting of three levels only. In fact, we would like to consider some real situation like an excited molecule but the computational burden imposed by the theoretical methods is so large, that we have been forced to carry out our comparisons on this simple model.

Sec. II.B.1 explains how the stationary Wiener- Khintchine spectrum can be calculated in the Schrödinger picture using the quantum regression theorem, this is used as a reference in the following. In Sec.II.C.1 it is shown how the quantum regression theorem can be used to obtain the physical spectrum of Eberly and Wódkiewicz. We will use these results to judge the reliability of the method presented in this paper. Sec. II.C.2 presents the theory of our own approach, which relies heavily on the work by Carmichael [11]. The derivation leads to a master equation, which is used in a quantum simulation to obtain spectra as explained above. The results are presented in Sec. III and compared with the physical spectra in Sec. III.C. Finally Sec. IV contains the conclusions.

II. MEASURING THE TIME-DEPENDENT SPECTRUM

A. The system under investigation

The model system we choose for our investigation is the 3-level atom shown in Fig. 1. The three levels are taken to have the (dimensionless) energies: $\omega_1 = 0$, $\omega_2 = 4.0$ and $\omega_3 = 8.0$. The levels $|2\rangle$ and $|3\rangle$ decay to level $|1\rangle$ by the rates $\Gamma_1 = \Gamma_2 = 0.1$. These parameters are kept constant all through the present paper. The level pairs $|2\rangle < - > |1\rangle$ and $|3\rangle < - > |2\rangle$ are coupled through lasers with the Rabi frequencies Ω_1 and Ω_2 respectevely. These coupling strengths are varied in the calculations. The fact, that the dipole approximation does not allow all transitions considered, lacks significance for the features we are investigating in this paper. In a molecular system parity may not be a good quantum number, in an atom or quantum dot system, the decay Γ_2 may derive from higher multipole transitions.
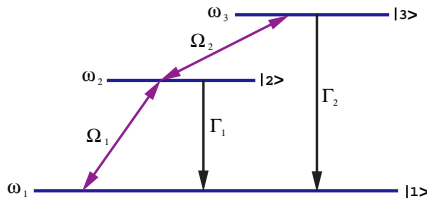

FIG. 1. The three level atom used in the calculations. The atom has three energy levels $\omega_1 = 0.0$, $\omega_2 = 4.0$ and $\omega_3 = 8.0$, two internal decays $\Gamma_1 = \Gamma_2 = 0.1$ and two lasers Ω_1 and Ω_2.

Mathematically the dynamics of this kind of system can be studied using master equations. We have included two different spontaneous decays to the reservoirs so the master equation has two decay terms of the Lindblad form. The reservoir is taken to be at zero temperature. Assuming the Born-Markov and the Rotating Wave Approximations (RWA) we find the master equation

$$\frac{d\hat{\varrho}}{dt} = -i[\hat{H}, \hat{\varrho}] + \sum_{i=1}^{2} \Gamma_i (\hat{L}_i \hat{\varrho} \hat{L}_i^\dagger - \frac{1}{2}\hat{L}_i^\dagger \hat{L}_i \hat{\varrho} - \frac{1}{2}\hat{\varrho}\hat{L}_i^\dagger \hat{L}_i) \quad (1)$$

where $\hat{L}_1 = |1\rangle\langle 2|$ and $\hat{L}_2 = |1\rangle\langle 3|$ are Lindblad operators. Everywhere in this paper the units have been chosen in such a way that $\hbar = 1$. Lasers are taken into account by adding resonant driving terms to the Hamiltonian; the laser frequencies are chosen such that

$$\omega_{l1} = \omega_2 - \omega_1,$$
$$\omega_{l2} = \omega_3 - \omega_2. \quad (2)$$

In the rotating-wave approximation the Hamiltonian becomes

$$\hat{H} = \sum_{i=1}^{3} \omega_i |i\rangle\langle i| + \frac{\Omega_1}{2}(|1\rangle\langle 2|e^{i\omega_{l1}t} + \text{h.c}) +$$
$$\frac{\Omega_2}{2}(|2\rangle\langle 3|e^{i\omega_{l2}t} + \text{h.c}). \quad (3)$$

B. The measurement scheme and the stationary spectrum

The usual setup for spectrum measurements is presented in Fig. 2. Fluorescence radiation from the three level atom is guided to the filter which allows radiation with only a certain frequency ω_D to go through it. Behind the filter, a photodetector detects the intensity of the radiation coming through. The spectrum can be measured by scanning ω_D over an appropriate frequency range and recording the relative intensities. The same measurement configuration can be used to measure both stationary and time-dependent spectra.
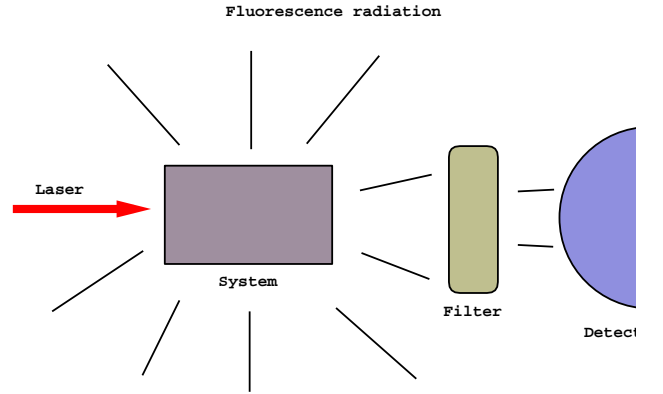

FIG. 2. The measurement scheme. The system is driven by a laser. Part of the fluorescence radiation passes the filter to the detector.

1. Stationary spectrum and the Quantum Regression Theorem

Mathematically the spectrum for a stationary system, the Wiener-Khintchine spectrum, is defined as

$$S_{WK}(\omega) = \int_{-\infty}^{\infty} d\tau \langle \hat{a}^\dagger(t)\hat{a}(t + \tau)\rangle e^{i\omega\tau}, \quad (4)$$

where $\hat{a}$ and $\hat{a}^\dagger$ are field operators. The definition contains two-time averages of the field modes. These expectation values are averages over the degrees of freedom of

the system, so in practise the spectrum is calculated from two-time system expectation values.

In the calculation of the spectrum, two-time averages of system operators in the Heisenberg picture are needed. Simulations are, however, done in the Schrödinger picture. The method which allows us to calculate multitime averages in the Heisenberg picture using a master equation is called the Quantum Regression Theorem (QRT).

The calculation of the expectation value $\langle \hat{O}_2(t + \tau)\hat{O}_1(t)\rangle$, $\tau \geq 0$ can be carried out with the QRT using the following steps:

1. Evolve the density matrix $\hat{\varrho}$ using the master equation from the initial time t_0 to the time t.

2. Form the matrix $C_{ij}(t, \tau = 0)$

$$C_{ij}(t, \tau = 0) = \text{Tr}[\varrho(t)|i\rangle\langle j|\hat{O}_1], \qquad (5)$$

where $\hat{O}_1$ is the operator in the Schrödinger picture.

3. Evolve $C_{ij}(t, \tau)$ using equation (1) with the initial matrix $C_{ij}(t, \tau = 0)$

$$\frac{dC_{ij}(t, \tau)}{d\tau} = G_{ijkl}C_{kl}(t, \tau); \qquad (6)$$

here repeated indices are summed over. Equation (6) is the master equation (1) in component form. The coefficients G_{ijkl} are thus determined by the master equation.

4. Form $\langle \hat{O}_2(t + \tau)\hat{O}_1(t)\rangle$ using the matrix elements of $\hat{O}_2$ in the Schrödinger picture ie.

$$\langle \hat{O}_2(t + \tau)\hat{O}_1(t)\rangle = \hat{O}_{2,ij}C_{ij}(t, \tau). \qquad (7)$$

This procedure gives the expectation values for such time pairs $(t, t + \tau)$ where the operator $\hat{O}_2$ has a larger time value than the operator $\hat{O}_1$, ie. $\tau \geq 0$. In order to get time values in the reverse order, we have to take a complex conjugate. It is also possible to calculate expectation values of the type $\langle \hat{O}_1(t)\hat{O}_2(t + \tau)\hat{O}_3(t)\rangle$. The difference is that at step 2 we have to calculate

$$C_{ij}(t, \tau = 0) = Tr[|i\rangle\langle j|\hat{O}_1\varrho(t)\hat{O}_3] \qquad (8)$$

and then continue as in the previous case.

2. The Stationary spectrum for the three level system

Without performing any calculations, we expect the following kind of structure of the stationary spectrum of spontaneous emission for the three level system. When the laser amplitudes are small, the spectrum should have two peaks whose relative intensities depend on the parameters Γ_1 and Γ_2. If Γ_1 is large, then only level two should decay significantly and the peak from transition from level three to one should be small. When the laser intensities are increased, we should see some kind on Rabi splitting in the spectrum, ie. the two peaks are expected to display substructure.

The detection operator used in the calculations is

$$\hat{O} = \Gamma_1|1\rangle\langle 2| + \Gamma_2|1\rangle\langle 3|. \qquad (9)$$

The two-time average $\langle \hat{O}^\dagger(t)\hat{O}(t+\tau)\rangle$ has been calculated and the spectrum is obtained by integrating this expectation values with respect to τ. In Figs. 3 and 4 the stationary spectrum of our three level system is shown for two different laser amplitudes. In Fig 3 the lasers are weak and we see two narrow peaks from the two transitions. In Fig. 4 the lasers are ten times stronger compared to the first case. The energy levels are split and the spectrum has got an eight peak structure, which can be understood as follows: Because of the strong lasers all three energy levels are split into sublevels. At low intensity we have observed two spectral components, at higher driving fields each one is seen to split into four subcomponents. The frequencies are different for all four transitions. This corresponds exactly to the number of degrees of freedom in a normalized 3×3 density matrix. The two time averages have an oscillating structure when the time separation τ is large. In practical calculations, infinities in equation (4) have been replaced by large time values. As a result we get asymmetries in the spectra and also finite linewidths.
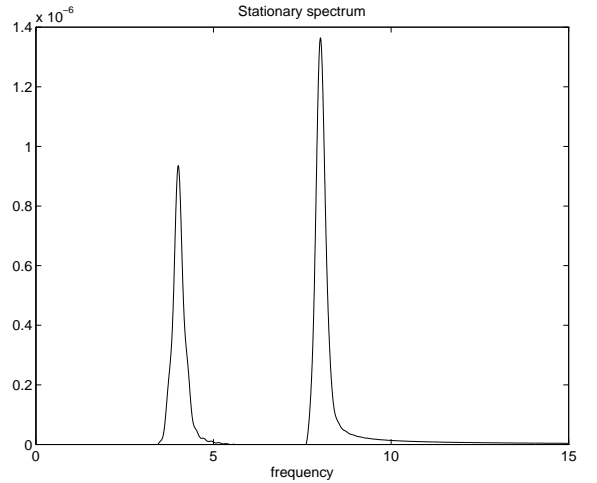

FIG. 3. The stationary spectrum for our three level atom when lasers are weak. The energy levels and decay constants are the same as in Fig. 1. The laser amplitudes are $\Omega_1 = \Omega_2 = 0.2$.

C. The physical spectrum

One weakness of the WK-spectrum is that it is only defined if the system is in a stationary state. This is clearly a limitation. For example, if instead of having a driving laser with constant intensity, we took a laser whose intensity changes in time, we obtain fluorescence
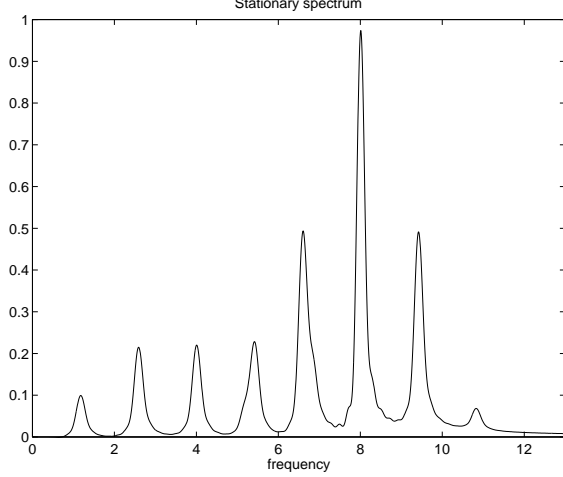
FIG. 4. The stationary spectrum for our three level atom when the lasers are strong. The energy levels and decay constants are the same as in Fig. 1. The laser amplitudes are $\Omega_1 = \Omega_2 = 2.0$.

light with varying intensity and spectrum. The fluorescence light displays a spectrum but we cannot calculate it using the WK-definition. There are several suggested definitions of a spectrum which would be physical also for nonstationary systems [1–3]. In the stationary limit all these spectra give the WK-spectrum.

A generalization which takes into account also the measurement scheme was proposed by Eberly and Wòdkiewicz [4]. What they call the physical spectrum, is defined as

$$S_{PHYS}(t, \omega_f, \Gamma_f) = \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 H^*(t - t_1, \omega_f, \Gamma_f) \cdot$$
$$H(t - t_2, \omega_f, \Gamma_f)\langle V^*(t_1)V(t_2)\rangle, \qquad (10)$$

where $H(t, \omega_f, \Gamma_f)$ is a filter function characteristic of the filter (see Fig.2)

$$H(t, \omega_f, \Gamma_f) = \int_{-\infty}^{\infty} d\omega H(w, \omega_f, \Gamma_f)e^{-i\omega t}d\omega. \qquad (11)$$

$H(\omega, \omega_f, \Gamma_f)$ is a function of ω, the parameter ω_f is the mean value of the passband of the filter and Γ_f is its width. The filter function of a Fabry-Perot filter, which is used in our calculations, has the form

$$H(t, \omega_f, \Gamma_f) = \Theta(t)\Gamma_f \exp(-(\Gamma_f + i\omega_f)t), \qquad (12)$$

which gives for physical spectrum the expression

$$S_{PHYS}(t, \omega_f, \Gamma_f) = \Gamma_f^2 \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t} dt_2$$

$$e^{-(\Gamma_f - i\omega_f)(t - t_1)}e^{-(\Gamma_f + i\omega_f)(t - t_2)}\langle V^*(t_1)V(t_2)\rangle =$$

$$\Gamma_f^2 \int_{0}^{\infty} d\tau_1 \int_{0}^{\infty} d\tau_2 e^{-(\Gamma_f - i\omega_f)\tau_1}e^{-(\Gamma_f + i\omega_f)\tau_2} \,.$$

$$\langle V^*(t - \tau_1)V(t - \tau_2)\rangle. \qquad (13)$$

From the definitions above, we see that as well as for the Wiener-Khintchine spectrum, two time averages are needed. The difference compared with S_{WK} is that now we need only correlation functions whose times are less than the time at which we calculate the spectrum.

1. Calculation of the physical spectrum using QRT

In order to calculate the physical spectrum at time T, we need to calculate the correlation functions $\langle \hat{O}^\dagger(t_1)\hat{O}(t_2)\rangle$, $0 \le t_1, t_2 \le T$. For numerical calculations we have to discretize the time $t_1, t_2 = n \cdot \Delta t$, $n = 0...N$, $\Delta t = \frac{T}{N}$. There are now N time instants in between $t = 0$ and $t = T$. The algorithm is the following:

1.At $t = 0$, form $C_{ij}(t_2 = 0, t_1 = 0)$ using the initial density matrix $\varrho(0)$ and use the algorithm presented in the last section to calculate $C_{ij}(t_2, t_1)$ for the time values $t_2 = 0$, $0 \le t_1 \le T$.

2. Evolve $\hat{\varrho}(t)$ to the time Δt, form $C_{ij}(\Delta t, t_1)$ and use QRT to evolve it to $t_1 = T$. This gives the correlations at $t_2 = \Delta t$, $\Delta t \le t_1 \le T$.

3. Evolve $\varrho(t)$ to the next time step, form C_{ij} with a new t_2 and evolve it to $t_1 = T$. Repeat this step until $t_2 = T$.

4. The algorithm above gives correlations for time pairs $t_2 = n \cdot \Delta t$, $t_2 \le t_1 \le T$, ie. $t_2 \le t_1$. In order to obtain results for time values in the reverse order, take the complex conjugate $C(t_1, t_2)^* = C(t_2, t_1)$.

Once we have calculated the correlation functions on a grid dense enough, the physical spectrum is a sum over the grid according to equation (10).

The detection operator used is the same as in the stationary case (9). Now the expectation values $\langle \hat{O}^\dagger(t_1)\hat{O}(t_2)\rangle$ are calculated using the algorithm for a nonstationary spectrum. Figures 5,6 and 7 show time dependent physical spectra calculated by the method described above. The parameters are the same as in Fig. 4, so we expect the steady state spectrum to show eight peaks. The initial state of the system is $\hat{\varrho}(0) = |2\rangle\langle 2|$. For the time dependent spectrum also the initial state has some influence. Spectra with different initial states are different for short times.

From Fig 5 figure it is seen that when time is small the spectrum has two broad peaks. At $t = 4.0$ a little more structure begins to appear, and at $t = 8.0$, eight peaks can be seen. When time increases the spectrum approaches the steady state spectrum, cf. Fig. 7. The broad spectrum features at small times derive from the uncertainty principle, which does not allow us to see too detailed structures of the spectrum, or more precisely,

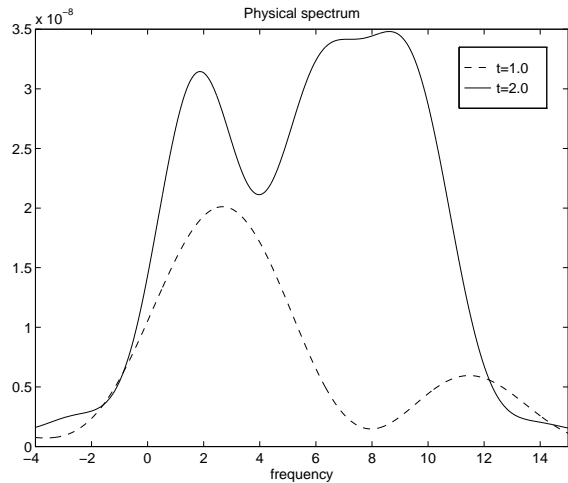the spectrum does not display any detailed structure at small time values.

FIG. 5. The time dependent physical spectrum calculated using QRT at the times $t = 1.0$ and $t = 2.0$. The parameters as in Fig. 4.

2. The theory of cascaded open system

Next we show how the theory of cascaded open systems [10,11] can be used to calculate the time dependent spectrum. By this we mean a system (B) which is driven by radiation coming from another quantum system (A). Radiation from the system A, which may be for example fluorescence light, is mediated through a reservoir to a system B. The coupling is uni-directional, the radiation from system B does not go to the system A. The system configuration is shown in Fig. 8. In the following, we derive a master equation for such a system, following the presentation in the paper by Carmichael [11].

The initial Hamiltonian is the following

$$\hat{H} = \hat{H}_A + \hat{H}_B + \hat{H}_R + \hat{H}_{AR} + \hat{H}_{BR}, \qquad (14)$$

where $\hat{H}_A$ and $\hat{H}_B$ are the Hamiltonians for systems A and B respectively. $\hat{H}_R$ is the reservoir Hamiltonian which mediates radiation from A to B. $\hat{H}_{AR}$ and $\hat{H}_{BR}$ are coupling Hamiltonians to the reservoir

$$\hat{H}_{AR} = i\sqrt{\gamma_A}(\hat{a}\hat{E}^\dagger(0) - \text{h.c})$$
$$\hat{H}_{BR} = i\sqrt{\gamma_B}(\hat{b}\hat{E}^\dagger(l) - \text{h.c}), \qquad (15)$$

where $\hat{a}$ and $\hat{b}$ are annihilation operator for system A and B respectively. The coupling to the reservoir is in two different spatial locations for the systems A and B. We next proceed to eliminate this complication. Using the unitary transformation

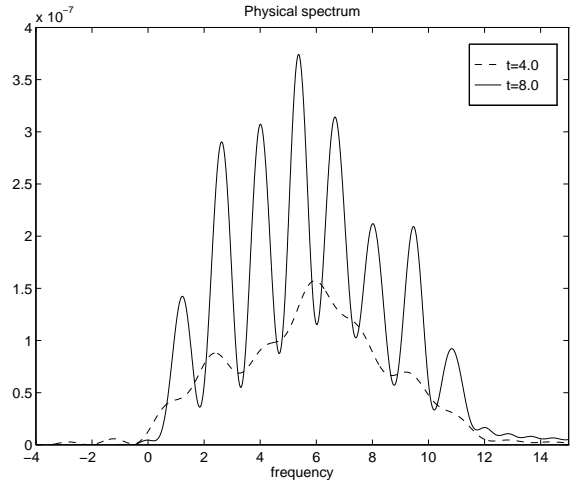$$\hat{U}_A(\tau) = \exp[i(\hat{H}_A + \hat{H}_R + \hat{H}_{AR})\tau], \qquad (16)$$

FIG. 6. The time dependent physical spectrum calculated using QRT at the times $t = 4.0$ and $t = 8.0$. The parameters the same as in Fig. 4.
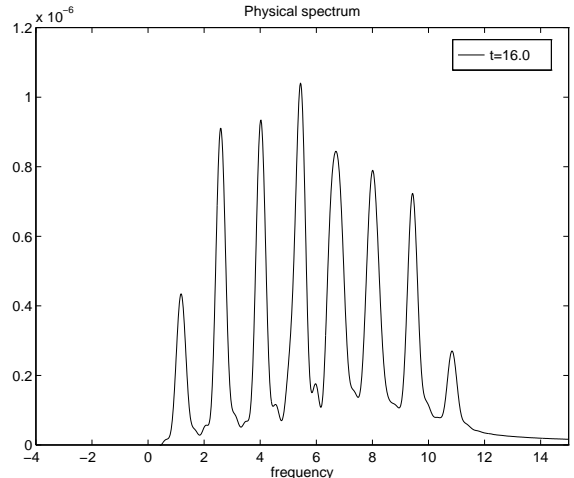

FIG. 7. The time dependent physical spectrum calculated using QRT at time $t = 16.0$. The parameters the same as in Fig. 4.

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we obtain an equation for the transformed density matrix $\hat{\varrho}'$ with Hamiltonians which are now in the standard form

$$\hat{H}' = \hat{H}_S + \hat{H}_R + \hat{H}_{SR}, \tag{17}$$

where

$$\hat{H}_S = \hat{H}_A + \hat{H}_B + \frac{i}{2}\sqrt{\gamma_A\gamma_B}(\hat{a}^\dagger\hat{b} - \text{h.c})$$
$$\hat{H}_{SR} = i(\sqrt{\gamma_A}\hat{a} + \sqrt{\gamma_B}\hat{b})(\hat{E}^\dagger(0) - \text{h.c}) \tag{18}$$

The master equation can now be derived in the conventional manner by adiabatic elimination of the reservoir. With these Hamiltonians the derivation gives

$$\frac{\partial\hat{\varrho}'}{\partial t} = -i[\hat{H}_S, \hat{\varrho}'] + \hat{C}\hat{\varrho}'\hat{C}^\dagger - \frac{1}{2}\hat{C}^\dagger\hat{C}\hat{\varrho}' - \frac{1}{2}\hat{\varrho}'\hat{C}^\dagger\hat{C}, \tag{19}$$

where the Lindblad operator $\hat{C}$ is

$$\hat{C} = \sqrt{\gamma_A}\hat{a} + \sqrt{\gamma_B}\hat{b}. \tag{20}$$

Next we give a short description how the theory described above can be used for spectrum measurements. As our detector system we choose a two level atom which is in the ground state. When we start to drive the atom using radiation with very low intensity, the excited state of the atom appears with a small probability. The probability of the excited state is a function of intensity and the spectrum of the radiation. If the atom has a very narrow linewidth, then only that part of the radiation which is in resonance can excite it. Because excitation is linearly proportional to the low intensity, the probability of the excited state is proportional to the the intensity of the incoming radiation at the atomic resonance frequency.
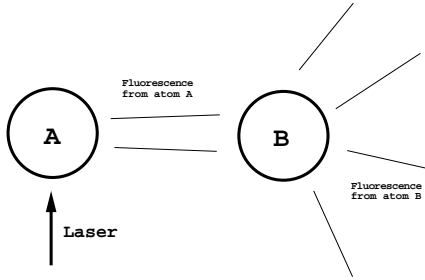

FIG. 8. The schematic layout of cascaded open systems. Fluorescence radiation from atom A goes to atom B, but the fluorescence radiation from B does not go back to the system A

III. CALCULATION OF THE TIME DEPENDENT SPECTRUM FOR THE THREE LEVEL ATOM

A. The master equation for spectrum analyzer

The configuration for a spectrum analyzer is shown in Fig. 9. The system B is a two level atom which is

used to analyze the radiation emitted from the system A. The atom B has a small decay constant Γ_B which corresponds to Γ_F in the definition of the physical spectrum (10). The system A is driven by a laser. Only a very small fraction of the fluorecent radiation coming from system A is guided to atom B ie. the constant p is very small. The majority of the radiation $(1-p)\Gamma_A$ goes into other directions and misses the detector. Division of the radiation is needed, because otherwise the incoming radiation would saturate the atom B and excitation would not be linearly proportional to the intensity any more. Figure 10 shows the probabilities of the excited state of an assembly of analyzer atoms as functions of time for the system described in detail later in this paper. Different curves correspond to different values of the resonance frequency. The spectrum is obtained by plotting the excitation probability at any given time with ω_B as a parameter. This method does not need to evaluate multitime averages as in all earlier definitions. The spectrum is obtained from one time averages of another quantum system, which is coupled to the system we are interested in. This method of spectrum calculation is quite close to the method presented in the book by M.Sargent III et.al. [12].

The system studied was described in section II. The time dependent spectrum of this three level atom has been calculated using a two level atom as a spectrum analyzer. The master equation for the three level atom and an analyzer atom according to the theory of cascaded open systems is the following.

$$\frac{\partial\hat{\varrho}}{\partial t} = -i[\hat{H}_A + \hat{H}_B + \hat{H}_C, \hat{\varrho}] +$$
$$\sum_{i=1}^{4}(\hat{L}_i\hat{\varrho}\hat{L}_i^\dagger - \frac{1}{2}\hat{L}_i^\dagger\hat{L}_i\hat{\varrho} - \frac{1}{2}\hat{\varrho}\hat{L}_i^\dagger\hat{L}_i), \tag{21}$$

where

$$\hat{H}_A = \sum_{i=1}^{3} E_i|i\rangle\langle i| + (|1\rangle\langle 2|e^{i(\omega_2 - \omega_1)t} + \text{h.c}) +$$
$$(|2\rangle\langle 3|e^{i(\omega_3 - \omega_2)t} + \text{h.c})$$
$$\hat{H}_B = \frac{1}{2}\omega_B\sigma_z \tag{22}$$
$$\hat{H}_C = \frac{i}{2}\sqrt{\gamma_1 p\Gamma_B}(|2\rangle\langle 1|\hat{\sigma}_- - \text{h.c}) +$$
$$\frac{i}{2}\sqrt{\gamma_2 p\Gamma_B}(|3\rangle\langle 1|\hat{\sigma}_- - \text{h.c})$$

and the Lindblad operators are

$$\hat{L}_1 = \sqrt{\Gamma_{A1}(1-p)}\,|1\rangle\langle 2|$$
$$\hat{L}_2 = \sqrt{\Gamma_{A2}(1-p)}\,|1\rangle\langle 3| \tag{23}$$
$$\hat{L}_3 = \sqrt{\Gamma_{A1}p}|1\rangle\langle 2| + \sqrt{0.5\Gamma_B}\,\hat{\sigma}_-^B$$
$$\hat{L}_4 = \sqrt{\Gamma_{A2}p}|1\rangle\langle 3| + \sqrt{0.5\Gamma_B}\,\hat{\sigma}_-^B.$$

In addition to the atomic Hamiltonians, there is an additional term in the Hamiltonian arising from the derivation which gives a small energy shift. The first two decay operators describe decays of the three level atom which take account of the majority of the radiation. They correspond to the $(1-p)\Gamma_A$ term in Fig. 9. The last two terms describe that part of the radiation which goes to the analyzer atom, the $p\Gamma_A$ term in Fig. 9. They also include spontaneous emission from atom B.

It can be seen that that if $p = 0$ the atoms are not coupled. Then, if the initial state is factorizable then the two atoms evolve separately. Atom A has two decay terms, which describe the transitions $|1\rangle\langle2|$ and $|1\rangle\langle3|$. The Analyzer atom has one decay term with the decay constant Γ_B. This separation is natural because if $p = 0$ there is no radiation coming from atom A to atom B. If p is nonzero it is possible to trace atom B out of the equations and we get the well known master equation for atom A. This means that system B does not affect the time evolution of system A at all.
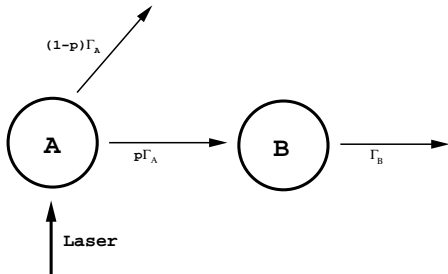

FIG. 9. This is the version of the cascaded open system we use as a time dependent spectral analyzer. A small portion p of radiation from atom A goes to atom B. The majority of the radiation goes into other directions $(1-p)$. The system B decays with decay constant Γ_B.

B. Simulations studied

The first case to be studied is our three level system with weak lasers. Figure 11 shows the time evolution of the diagonal elements of the density matrix. The initial state is $\hat{\varrho}(0) = |2\rangle\langle2|$. At first the population of level two decreases and starts to Rabi oscillate, at large times it approaches a constant and reaches its steady state. The populations of levels one and three increase in the beginning and for large times reach steady state. The behaviour of the off-diagonal elements is quite similar (Fig. 12), first there are oscillations which smoothen out at large times. In Fig. 10 the population of the excited level is shown for all 128 analyzer atoms used in our simulations. The different curves belong to different analyzer atoms with different frequencies. They are clearly excited

differently and this means that the atoms can really recognize the spectral structure of the incoming radiation. The simulations were done using the Monte-Carlo wavefunction technique [9,13–15]. The curves are not very smooth so bigger ensembles would be needed. The numerical calculations were, however, very demanding and 128 different target atoms and the size of their ensembles were chosen for practical reasons.

Spectra read from the analyzer atoms with weak lasers are shown in Fig. 13, 14 and 15. The parameters are the same as in Fig 3. The parameter p in the Lindblad operators (23) is chosen to be $p = 0.005$. The decay constant for analyzer atoms is $\Gamma_B = 0.001$. The initial state is $\hat{\varrho}(0) = |2\rangle\langle2|$.

In figure 13 the spectrum is broad. At $t = 2.0$ there is a broad peak centered around $\omega = 4$ and as time increases the peak becomes narrower which can be seen at $t = 8.0$ (Fig. 14). The appearance of a peak around $\omega = 4$ is understandable, because initially level two was populated. The spectrum reveals that the atom decays from level two to the ground level. As time increases, another peak begins to appear at $\omega = 8.0$. Its relative intensity increases and in steady state it is bigger than the $\omega = 4.0$ peak. At long times, $t = 200$, the spectrum reaches its steady state Fig. 15; cf. Fig 3.

The spectra with stronger lasers are shown in figures 16,17 and 18. At small times, the spectrum is broad and the more precise structures appears when time increases. In the last figure the time is very large $t = 200$ and spectrum has reached its steady state Fig. 4.
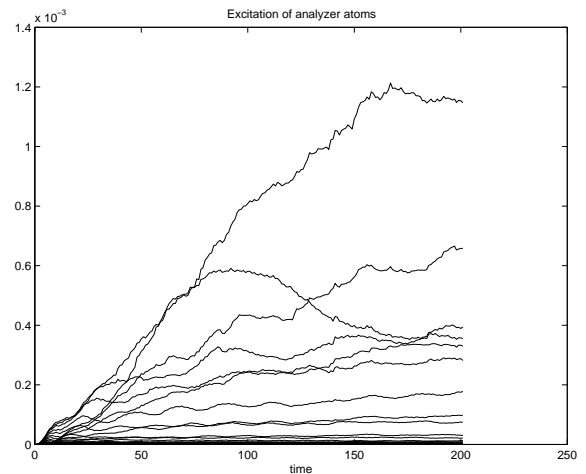

FIG. 10. Excitation of all 128 analyzer two-level atoms as functions on time. For most of the atoms, the excitations is so small that it cannot be seen. The three level system parameters are the same as in Fig. 1. The laser amplitudes are $\Omega_1 = \Omega_2 = 2.0$, the parameter $p = 0.005$, and the decay constant of the analyzer atoms $\Gamma_B = 0.001$. Each curve in this figure is obtained from an ensemble of identical atoms with 300 members.
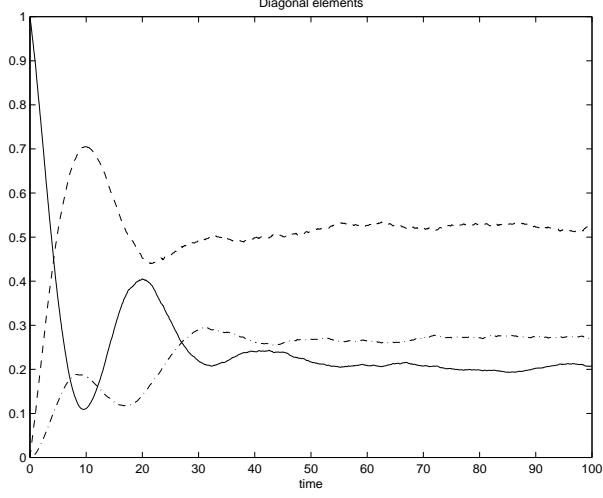
FIG. 11. Time evolution of diagonal elements. The energy levels and decay constants are the same as in Fig. 1. The laser amplitudes are $\Omega_1 = \Omega_2 = 2.0$. Dashed line: ϱ_{11}, solid line: ϱ_{22}, dashdot line: ϱ_{33}.
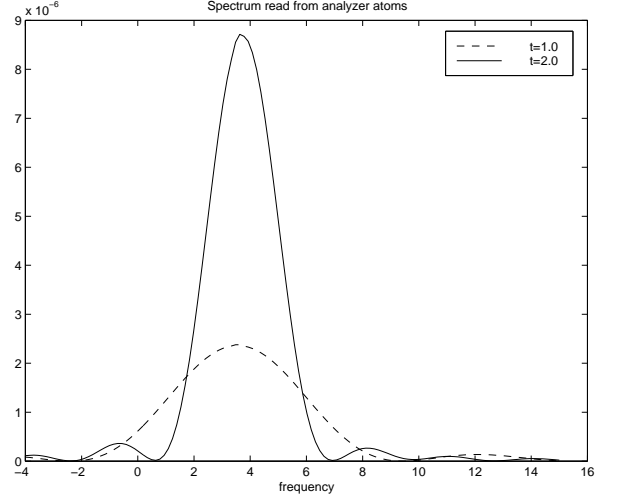

FIG. 13. The time dependent spectrum read from the analyzer atoms at times $t = 1.0$ and $t = 2.0$. The parameters are the same as in Fig. 3.
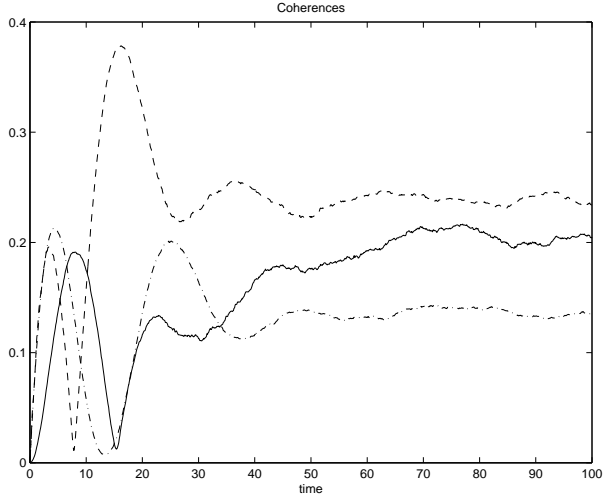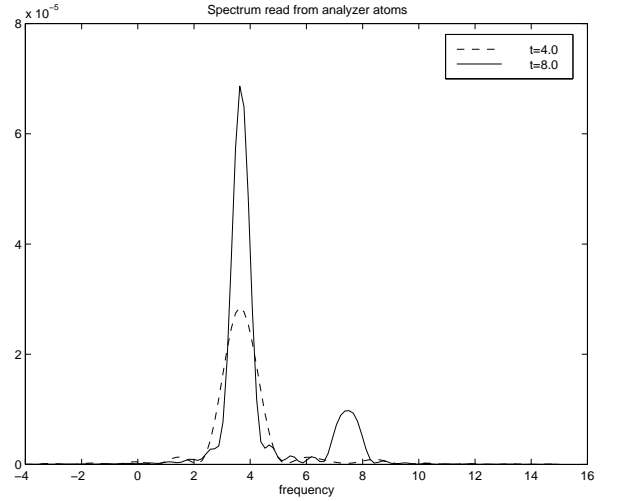

FIG. 12. Time evolution of absolute values of off-diagonal elements. The parameters are the same as in Fig. 11. Dashed line: $|\varrho_{12}|$, solid line: $|\varrho_{13}|$, dashdot line: $|\varrho_{23}|$.

FIG. 14. The time dependent spectrum read from the analyzer atoms at times $t = 4.0$ and $t = 8.0$. The parameters are the same as in Fig. 3.
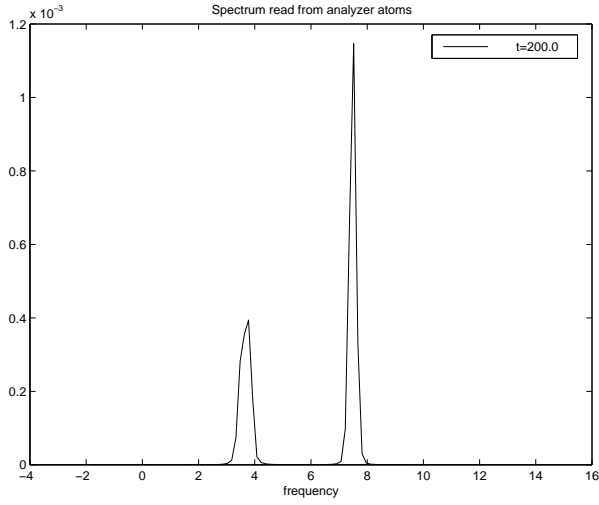
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FIG. 15. The time dependent spectrum read from the analyzer atoms at time $t = 200.0$. The parameters are the same as in Fig. 3.
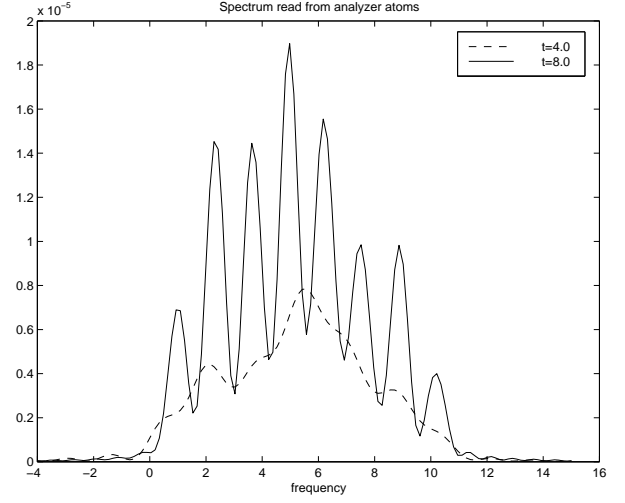

FIG. 17. The time dependent spectrum read from the analyzer atoms at times $t = 4.0$ and $t = 8.0$. The parameters are the same as in Fig. 4. Compare this with Fig. 6 calculated with the same parameters.
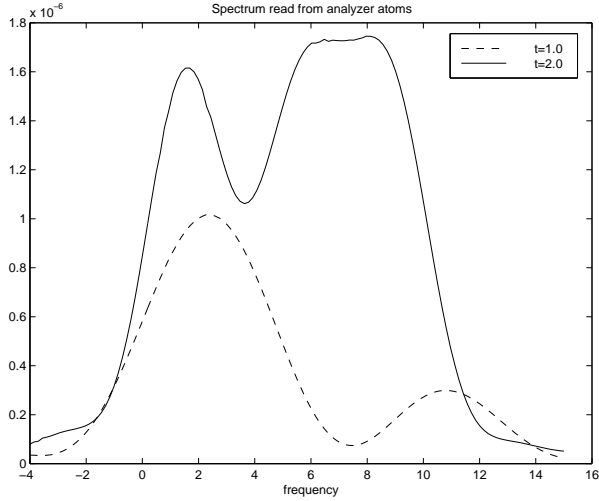

FIG. 16. The time dependent spectrum read from the analyzer atoms at times $t = 1.0$ and $t = 2.0$. The parameters are the same as in Fig. 4. Compare this with Fig. 5 calculated with the same parameters.
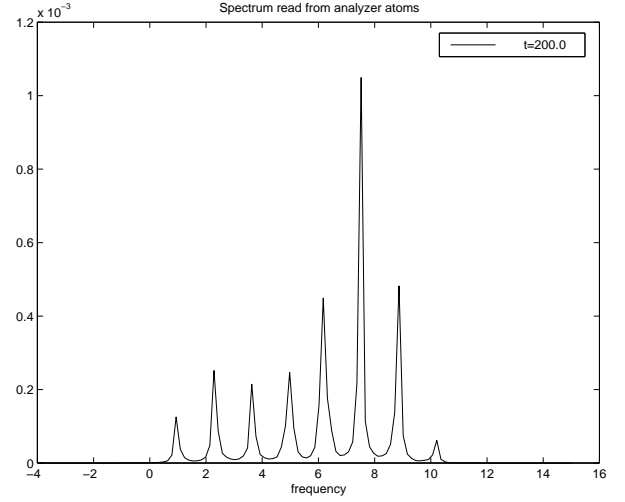

FIG. 18. The time dependent spectrum read from the analyzer atoms at time $t = 200.0$. The parameters are the same as in Fig. 4. Compare this with Fig. 4 calculated with the same parameters.

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C. Comparison between methods

The spectrum calculated using analyzer atoms and the definition of the physical spectrum seem to give very similar results. However, the calculation techniques are totally different and from the mathematics it is not at all obvious that these two spectra should agree. For the physical spectrum, two time averages are needed and the spectrum is a kind of Fourier-transform of them. In this respect the physical spectrum is closer to the Wiener-Khintchine spectrum. The method with analyzer atoms does not need any multitime-averages. The spectrum is read from a quantum system (the two state atom) which acts as a measurement device.

It is also interesting to compare how the uncertainty principle restricts spectral measurements. In all our simulations, analyzer atoms of very small linewidth were used. This means that the atoms respond to the changes of the incoming radiation very slowly, but if the interaction time is long enough the atoms give an accurate spectrum. If we used atoms with a larger linewidth, the atoms would indicate the state of the system more quickly, but because of their large linewidth they would not give a very accurate spectrum. In the case of a the physical spectrum, the uncertainty relation is related to the properties of the Fourier transform.

The method of analyzer atoms is based on the simulation of a master equation. Recently many parallel stochastic algorithms have been presented [11,13–15]. There are also freely available programs which are developed for simulations of these kinds of master equations [16]. It is also possible to parallelize the algorithm by computing the evolution of the different analyzer atoms on different processors.

IV. CONCLUSION

We have shown a fundamentally new method to calculate a spectrum. Instead of calculating two-time averages using the Quantum Regression Theorem we couple the system to two state atoms which are used as a measurement device. The spectrum is read from the excitation probability ie. from one-time averages of the two state atoms. This method allows us to calculate both the time dependent and stationary spectrum. Comparisons between the spectra calculated show that using this new method we find similar results as using two-time averages and a Fourier-transform.

Because of the frequency-time uncertainty, the spectral resolution is poor for initial times. However, when the bare level structure is considerebly modified by the presence of strong laser fields like in Fig. 4, it may not be possible to surmise the low intensity structure, Fig. 3, which is characteristic of the system of interest. The the short time spectra, Figs. 5 and 16 may indicate the number of transitions involved even if their positions (4 and 8 in our case) are very difficult to locate. When the measurement goes on, more data are collected and the spectral information approaches the steady state as closely as we desire for a meaningful measurement.

V. ACKNOWLEDGEMENTS

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